

C E R T I F I C A T I O N

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[CLAIMS]

[Claim 1]

5 A method for preparing a rare earth permanent magnet material comprising the steps of:

forming a sintered magnet form of a R^1 -Fe-B composition wherein R^1 is one or more elements selected from among rare earth elements inclusive of Y and Sc to a shape having a dimension of up to 20 mm along its maximum side and
10 a dimension of up to 2 mm in a magnetic anisotropy direction,

disposing a powder having an average particle size of up to 100 μ m comprising one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4
15 wherein R^2 , R^3 and R^4 each are one or more elements selected from among rare earth elements inclusive of Y and Sc on the sintered magnet form in a magnet-surrounding space within a distance of 1 mm from the surface of the magnet form and at an average filling factor of at least 10%, and

20 heat treating the magnet form and the powder at a temperature equal to or below the sintering temperature of the magnet in vacuum or in an inert gas for 1 minute to 10 hours, thereby absorbing at least one of R^2 , R^3 and R^4 contained in the powder into the sintered magnet.

25 [Claim 2]

A method for preparing a rare earth permanent magnet material according to claim 1, further comprising, after the absorption treatment, effecting aging treatment at lower than the temperature of the heat treatment.

30 [Claim 3]

A method for preparing a rare earth permanent magnet material according to claim 1 or 2, wherein in said one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 wherein R^2 , R^3 and R^4 each are one or
35 more elements selected from among rare earth elements inclusive of Y and Sc, R^2 , R^3 or R^4 contains at least 20 atom% of Dy and/or Tb.

[Claim 4]

A method for preparing a rare earth permanent magnet material according to claim 1, 2 or 3, wherein said powder comprising one or more members selected from an oxide of R^2 ,
5 a fluoride of R^3 , and an oxyfluoride of R^4 wherein R^2 , R^3 and R^4 each are one or more elements selected from among rare earth elements inclusive of Y and Sc and having an average particle size of up to 100 μm is disposed in the surface of the magnet form as a slurry thereof dispersed in an aqueous
10 or organic solvent.

[Claim 5]

A method for preparing a rare earth permanent magnet material according to any one of claims 1 to 4, wherein the sintered magnet form machined into the above shape is
15 cleaned with at least one of alkalis, acids and organic solvents, before the powder treatment.

[Claim 6]

A method for preparing a rare earth permanent magnet material according to any one of claims 1 to 4, wherein a
20 surface layer of the sintered magnet form machined into the above shape is removed by shot blasting, before the powder treatment.

[Claim 7]

A method for preparing a rare earth permanent magnet
25 material according to any one of claims 1 to 6, wherein cleaning with at least one of alkalis, acids and organic solvents, grinding, or plating or painting is carried out as a final treatment.

2004-304543

[SPECIFICATION]

[TITLE OF THE INVENTION] Preparation of Rare Earth Permanent
Magnet Material

5 [TECHNICAL FIELD]

[0001]

This invention relates to a method for preparing a
R-Fe-B system permanent magnet in which the coercive force
of a sintered magnet form is enhanced while controlling a
10 decline of its remanence; and more particularly, to a method
for preparing a high performance, compact or thin rare earth
permanent magnet material.

[BACKGROUND ART]

[0002]

15 Nd-Fe-B system permanent magnets have a growing range
of application due to their excellent magnetic properties.
While electronic equipment having magnets built therein
including computer-related equipment, CD players, DVD
players, and mobile phones are currently under the trend
20 toward size and weight reductions, higher performance and
energy saving, there exists a demand to enhance the
performance of Nd-Fe-B magnets, especially compact or thin
Nd-Fe-B sintered magnets.

[0003]

25 Indexes for the performance of magnets include
remanence (or residual magnetic flux density) and coercive
force. An increase in the remanence of Nd-Fe-B sintered
magnets can be achieved by increasing the volume factor of
Nd₂Fe₁₄B compound and improving the crystal orientation. To
30 this end, a number of modifications have been made on the
process. With respect to the increased coercive force,
among different approaches including grain refinement, the
use of alloy compositions with greater Nd contents, and the
addition of effective elements, the currently most common
35 approach is to use alloy compositions having Dy or Tb
substituted for part of Nd. Substituting these elements for
Nd in the Nd₂Fe₁₄B compound increases both the anisotropic

magnetic field and the coercive force of the compound. The substitution with Dy or Tb, on the other hand, reduces the saturation magnetic polarization of the compound.

Therefore, as long as the above approach is taken to
5 increase coercive force, a loss of remanence is unavoidable.

[0004]

In Nd-Fe-B magnets, the coercive force is given by the magnitude of an external magnetic field which creates nuclei of reverse magnetic domains at grain boundaries. Formation
10 of nuclei of reverse magnetic domains is largely dictated by the structure of the grain boundary in such a manner that any disorder of grain structure in proximity to the boundary invites a disturbance of magnetic structure, helping formation of reverse magnetic domains. The inventors found
15 that by concentrating trace Dy or Tb only in proximity to the grain boundaries to increase the anisotropic magnetic field only in proximity to the boundaries, the coercive force can be increased while suppressing significant decline of remanence (Patent Document 1). Subsequently, the
20 inventors established a production method comprising separately preparing a $\text{Nd}_2\text{Fe}_{14}\text{B}$ compound composition alloy and a Dy or Tb-rich alloy, mixing them and sintering the mixture (Patent Document 2). In this method, the Dy or Tb-rich alloy becomes a liquid phase during the sintering and is
25 distributed so as to surround the $\text{Nd}_2\text{Fe}_{14}\text{B}$ compound. As a consequence, substitution of Dy or Tb for Nd occurs only in proximity to grain boundaries in the compound, so that the coercive force can be effectively increased while suppressing significant decline of remanence.

30 [0005]

On the other hand, it is reported for small magnets that coercive force can be increased by applying Dy or Tb on the magnet surface by sputtering, and heat treating the magnet, thereby causing Dy or Tb to diffuse only to grain
35 boundaries (Non-Patent Documents 1 and 2). This method allows for more effective concentration of Dy or Tb at the grain boundary and succeeds in increasing the coercive force

without a substantial loss of remanence. As the magnet becomes larger in specific surface area, that is, the magnet form becomes smaller, the amount of Dy or Tb fed becomes larger, indicating that this method is applicable to only compact or thin magnets. However, there is still left the problem of poor productivity associated with the deposition of metal coating by sputtering or the like.

[Patent Document 1] JP-B 5-31807

[Patent Document 2] JP-A 5-21218

- 10 [Non-Patent Document 1] K.T. Park, K. Hiraga and M. Sagawa, "Effect of Metal-Coating and Consecutive Heat Treatment on Coercivity of Thin Nd-Fe-B Sintered Magnets," Proceedings of the Sixteen International Workshop on Rare-Earth Magnets and Their Applications, Sendai, p.257 (2000)
- 15
- 20 [Non-Patent Document 2] K. Machida, H. Kawasaki, M. Ito and T. Horikawa, "Grain Boundary Tailoring of Nd-Fe-B Sintered Magnets and Their Magnetic Properties," Proceedings of the 2004 Spring Meeting of the Powder & Powder Metallurgy Society, p.202
- 25

[DISCLOSURE OF THE INVENTION]

[Problem to be Solved by the Invention]

30 [0006]

An object of the invention, which is made to solve the foregoing problems, is to provide a method for preparing a compact or thin R¹-Fe-B sintered magnet having a high coercive force at a high productivity wherein R¹ is one or more elements selected from among rare earth elements inclusive of Y and Sc.

35

[Means for Solving the Problem]

[0007]

The inventors have discovered that when a R^1 -Fe-B sintered magnet, typically a Nd-Fe-B sintered magnet is
5 heated with a powder comprising one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 wherein R^2 , R^3 and R^4 each are one or more elements selected from among rare earth elements inclusive of Y and Sc being present in the magnet surface, R^2 , R^3 or R^4 contained
10 in the powder is absorbed in the magnet form whereby the coercive force is increased while significantly suppressing a decline of remanence. The present invention is predicated on this discovery.

[0008]

15 The present invention provides a method for preparing a rare earth permanent magnet material as defined below.

(1) A method for preparing a rare earth permanent magnet material comprising the steps of:

forming a sintered magnet form of a R^1 -Fe-B composition
20 wherein R^1 is one or more elements selected from among rare earth elements inclusive of Y and Sc to a shape having a dimension of up to 20 mm along its maximum side and a dimension of up to 2 mm in a magnetic anisotropy direction,

disposing a powder having an average particle size of
25 up to 100 μ m comprising one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 wherein R^2 , R^3 and R^4 each are one or more elements selected from among rare earth elements inclusive of Y and Sc on the sintered magnet form in a magnet-surrounding space within a
30 distance of 1 mm from the surface of the magnet form and at an average filling factor of at least 10%, and

heat treating the magnet form and the powder at a temperature equal to or below the sintering temperature of the magnet in vacuum or in an inert gas for 1 minute to 10
35 hours, thereby absorbing at least one of R^2 , R^3 and R^4 contained in the powder into the sintered magnet.

(2) A method for preparing a rare earth permanent magnet material according to (1), further comprising, after the absorption treatment, effecting aging treatment at lower than the temperature of the heat treatment.

5 (3) A method for preparing a rare earth permanent magnet material according to (1) or (2), wherein in said one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 wherein R^2 , R^3 and R^4 each are one or more elements selected from among rare earth elements
10 inclusive of Y and Sc, R^2 , R^3 or R^4 contains at least 20 atom% of Dy and/or Tb.

(4) A method for preparing a rare earth permanent magnet material according to (1), (2) or (3), wherein said powder comprising one or more members selected from an oxide of R^2 ,
15 a fluoride of R^3 , and an oxyfluoride of R^4 wherein R^2 , R^3 and R^4 each are one or more elements selected from among rare earth elements inclusive of Y and Sc and having an average particle size of up to 100 μm is disposed in the surface of the magnet form as a slurry thereof dispersed in an aqueous
20 or organic solvent.

(5) A method for preparing a rare earth permanent magnet material according to any one of (1) to (4), wherein the sintered magnet form machined into the above shape is cleaned with at least one of alkalis, acids and organic
25 solvents, before the powder treatment.

(6) A method for preparing a rare earth permanent magnet material according to any one of (1) to (4), wherein a surface layer of the sintered magnet form machined into the above shape is removed by shot blasting, before the powder
30 treatment.

(7) A method for preparing a rare earth permanent magnet material according to any one of (1) to (6), wherein cleaning with at least one of alkalis, acids and organic solvents, grinding, or plating or painting is carried out as
35 a final treatment.

[Benefits of the Invention]

[0009]

The present invention ensures that a compact or thin permanent magnet having a high remanence and a high coercive
5 force is manufactured at a high productivity.

[BEST MODE FOR CARRYING OUT THE INVENTION]

[0010]

Now the invention will be described in more detail.

The invention pertains to a method for preparing a
10 compact or thin R^1 -Fe-B sintered magnet material having a high remanence and coercive force.

[0011]

The R^1 -Fe-B sintered magnet form may be obtained from a mother alloy in a conventional way by coarse pulverization,
15 fine pulverization, compacting and sintering.

[0012]

The mother alloy contains R^1 , Fe, and B. R^1 represents one or more elements selected from among rare earth elements inclusive of Y and Sc, examples of which include Y, Sc, La,
20 Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Yb, and Lu. Preferably R^1 is mainly composed of Nd, Pr, and Dy. The rare earth elements inclusive of Y and Sc should preferably account for 10 to 12 atom%, especially 12 to 15 atom% of the entire alloy. More preferably, R^1 should contain either one
25 or both of Nd and Pr in an amount of at least 10 atom%, especially at least 50 atom%. Boron should preferably account for 3 to 15 atom%, especially 4 to 8 atom% of the entire alloy. The alloy may further contain 0 to 11 atom%, especially 0.1 to 5 atom% of one or more elements selected
30 from among Al, Cu, Zn, In, Si, P, S, Ti, V, Cr, Mn, Ni, Ga, Ge, Zr, Nb, Mo, Pd, Ag, Cd, Sn, Sb, Hf, Ta, and W. The balance consists of Fe and incidental impurities such as C, N and O. Iron should preferably account for at least 50 atom%, especially at least 65 atom% of the entire alloy. It
35 is acceptable that Co substitutes for part of Fe, for example, 0 to 40 atom%, especially 0 to 15 atom% of Fe.

[0013]

The mother alloy is obtained by melting the starting metals or alloys in vacuum or in an inert gas, preferably Ar atmosphere, and then pouring in a flat mold or book mold, or
5 pouring as by strip casting. An alternative method, called binary alloys method, is also applicable wherein an alloy whose composition is approximate to the $R_2Fe_{14}B$ compound, the primary phase of the present alloy and an R-rich alloy serving as a liquid phase aid at the sintering temperature are separately
10 prepared, crushed, weighed and admixed together. It is noted that since the alloy whose composition is approximate to the primary phase composition is likely to leave α -Fe depending on the cooling rate during the casting or the alloy composition, it is subjected to homogenizing treatment, if desired for the
15 purpose of increasing the amount of $R_2Fe_{14}B$ compound phase. The homogenization is achievable by heat treatment in vacuum or in an Ar atmosphere at 700 to 1,200°C for at least 1 hour. For the R-rich alloy serving as a liquid phase aid, not only the casting method described above, but also the so-called melt
20 quenching method are applicable.

[0014]

The alloy is generally coarsely pulverized to a size of 0.05 to 3 mm, especially 0.05 to 1.5 mm. For the coarse pulverizing step, a Brown mill or hydriding pulverization is
25 used, with the hydriding pulverization being preferred for the alloy prepared by strip casting. The coarse powder is then finely pulverized to a size of 0.2 to 30 μm , especially 0.5 to 20 μm , for example, on a jet mill using high pressure nitrogen. The fine powder is compacted in a magnetic field
30 by a compression molding machine and introduced into a sintering furnace. The sintering is carried out in vacuum or an inert gas atmosphere, typically at 900 to 1,250°C, especially 1,000 to 1,100°C.

[0015]

35 The sintered magnet thus obtained contains 60 to 99% by volume, preferably 80 to 98% by volume of the tetragonal

$R_2Fe_{14}B$ compound as the primary phase, with the balance being 0.5 to 20% by volume of an R-rich phase, 0 to 10% by volume of a B-rich phase, and at least one of carbides, nitrides, oxides and hydroxides resulting from incidental impurities or a mixture or composite thereof.

[0016]

The sintered block is machined into a preselected shape. In the practice of the invention, the shape should preferably have a dimension of up to 20 mm along its maximum side and a dimension of up to 2 mm in a magnetic anisotropy direction, for the reason that the amount of R^2 , R^3 or R^4 absorbed in the magnet form from the powder comprising one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 disposed on the magnet surface becomes greater as the specific surface area of the magnet form is larger, that is, the dimensions thereof are smaller. More preferably, the dimension in a magnetic anisotropy direction should be up to 1 mm.

[0017]

Disposed on the surface of the machined magnet form is a powder comprising one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 . It is noted that R^2 , R^3 and R^4 each are one or more elements selected from among rare earth elements inclusive of Y and Sc and preferably contains at least 20 atom%, more preferably at least 40 atom% of Dy or Tb. For the reason that a more amount of R^2 , R^3 or R^4 is absorbed as the filling factor of the powder in the magnet surface-surrounding space is higher, the filling factor should be at least 10% by volume, preferably at least 40% by volume, calculated as an average value in the magnet surrounding space from the magnet surface to a distance of 1 mm, in order for the invention to attain its effect. One exemplary technique of disposing or applying the powder is by dispersing a fine powder comprising one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 in water or an organic solvent to form a slurry, immersing the magnet

form in the slurry, and drying in hot air or in vacuum or drying in the ambient air. Alternatively, the powder can be applied by spray coating or the like. Any such technique is characterized by ease of application and mass treatment.

5 The particle size of the fine powder affects the reactivity when the R^2 , R^3 or R^4 component in the powder is absorbed in the magnet. Smaller particles offer a larger contact area that participates in the reaction. In order for the invention to attain its effect, the powder disposed around
10 the magnet should desirably have an average particle size of up to 100 μm , preferably up to 10 μm . It is noted that the average particle size is determined as a weight average diameter D_{50} (particle diameter at 50% by weight cumulative, or median diameter) upon measurement of particle size
15 distribution by laser light diffractometry.

[0018]

The oxide of R^2 , fluoride of R^3 , and oxyfluoride of R^4 used herein are typically $R^2_2O_3$, R^3F_3 , and R^4OF , respectively, although they generally refer to oxides containing R^2 and
20 oxygen, fluorides containing R^3 and fluorine, and oxyfluorides containing R^4 , oxygen and fluorine, additionally including R^2O_n , R^3F_n , and $R^4O_mF_n$ wherein m and n are arbitrary positive numbers, and modified forms in which part of R^2 , R^3 or R^4 is substituted or stabilized with another metal element
25 as long as they can achieve the benefits of the invention.

[0019]

After the powder comprising the oxide of R^2 , fluoride of R^3 , oxyfluoride of R^4 or a mixture thereof is disposed on the magnet surface, the magnet and the powder are heat
30 treated in vacuum or in an atmosphere of an inert gas such as argon (Ar) or helium (He). This treatment is referred to as absorption treatment, hereinafter. The temperature of absorption treatment is equal to or below the sintering temperature of the magnet form. The treatment temperature
35 is limited for the following reason.

[0020]

If treatment is done at a temperature above the sintering temperature (designated T_s in °C) of the relevant sintered magnet, there arise problems like (1) the sintered magnet alters its structure and fails to provide excellent magnetic properties; (2) the sintered magnet fails to maintain its dimensions as machined due to thermal deformation; and (3) the diffusing R can diffuse into the interior of magnet grains beyond the grain boundaries in the magnet, resulting in a reduced remanence. The treatment temperature should thus be equal to or below the sintering temperature, and preferably equal to or below $(T_s - 10)^\circ\text{C}$. The lower limit of temperature, which may be selected as appropriate, is typically at least 350°C . The time of absorption treatment is from 1 minute to 10 hours. The absorption treatment is not completed within less than 1 minutes whereas more than 10 hours of treatment gives rise to the problems that the sintered magnet alters its structure and the inevitable oxidation and evaporation of components adversely affect the magnetic properties. The more preferred time is 5 minutes to 8 hours, especially 10 minutes to 6 hours.

[0021]

Through the absorption treatment described above, R^2 , R^3 or R^4 which has been contained in the powder disposed on the magnet surface is transferred and concentrated in the R^1 -rich grain boundary phase component within the magnet, whereby the coercive force of the R^1 -Fe-B sintered magnet is effectively increased.

[0022]

In the absorption treatment, the magnets charged in a container are covered with the powder so that the magnets are kept apart, preventing the magnets from being fused together after the absorption treatment albeit high temperature. Additionally, the powder is not bonded to the magnets after the heat treatment. This permits a number of magnets to be placed in the container therein, indicating

that the preparation method of the invention is also improved in productivity.

[0023]

Also preferably, the absorption treatment is followed
5 by aging treatment. The aging treatment is desirably at a temperature which is below the absorption treatment temperature, preferably from 200°C to a temperature lower than the absorption treatment temperature by 10°C, and more preferably from 350°C to a temperature lower than the
10 absorption treatment temperature by 10°C. The atmosphere is preferably vacuum or an inert gas such as Ar or He. The time of aging treatment is from 1 minute to 10 hours, preferably from 10 minutes to 5 hours, and more preferably from 30 minutes to 2 hours.

15 [0024]

It is noted for the machining of the sintered magnet form that if the coolant used in the machining tool is aqueous, or if the surface being machined is exposed to high temperature during the machining, there is a likelihood of
20 an oxide film forming on the machined surface, which oxide film can inhibit the absorption reaction of R^2 , R^3 or R^4 component from the powder to the magnet form. In such a case, the oxide film is removed by cleaning with at least one of alkalis, acids and organic solvents or by shot
25 blasting before adequate absorption treatment is carried out.

[0025]

Examples of the alkali used herein include potassium pyrophosphate, sodium pyrophosphate, potassium citrate,
30 sodium citrate, potassium acetate, sodium acetate, potassium oxalate, sodium oxalate, etc. Examples of the acid used herein include hydrochloric acid, nitric acid, sulfuric acid, acetic acid, citric acid, tartaric acid, etc. Examples of the organic solvent used herein include acetone,
35 methanol, ethanol, isopropyl alcohol, etc. Herein, the alkali or acid may be used as an aqueous solution having an appropriate concentration not to attack the magnet form.

The magnet, which has been subjected to the absorption treatment and optionally, subsequent aging treatment, may be again cleaned with at least one of alkalis, acids and organic solvents or machined to a practical shape.

5 Moreover, the process including absorption treatment, aging treatment, and cleaning or machining may further include subsequent plating or painting.

[0026]

10 The permanent magnet material thus obtained finds use as a high-performance, compact or thin permanent magnet having an increased coercive force.

[EXAMPLES]

[0027]

15 Examples are given below for illustrating the present invention, but the scope of the invention is not limited thereby. In Examples, the filling factor of dysprosium oxide or dysprosium fluoride in the magnet surface-surrounding space is calculated from a weight gain of the magnet after powder treatment and the true density of powder material.

[0028]

[Example 1 and Comparative Examples 1-2]

25 A thin plate of alloy was prepared by a so-called strip casting technique, specifically by weighing predetermined amounts of Nd, Co, Al and Fe metals having a purity of at least 99% by weight and ferroboration, induction heating in an argon atmosphere for melting, and casting the alloy melt on a copper single roll in an argon atmosphere. The resulting alloy had a composition of 13.5 atom% Nd, 1.0 atom% Co, 0.5 atom% Al, 5.8 atom% B, and the balance of Fe and is designated Alloy A. Alloy A was hydrided and then heated at 500°C for partial dehydriding while evacuating to vacuum. By this so-called hydriding pulverization, the alloy was pulverized into a coarse powder having a size of up to 30 mesh. Another alloy was prepared by weighing 35 predetermined amounts of Nd, Tb, Fe, Co, Al and Cu metals having a purity of at least 99% by weight and ferroboration,

induction heating in an argon atmosphere for melting, and casting. The resulting alloy had a composition of 20 atom% Nd, 10 atom% Tb, 24 atom% of Fe, 6 atom% B, 1 atom% of Al, 2 atom% of Cu, and the balance of Co and is designated Alloy B. Using a Brown mill in a nitrogen atmosphere, Alloy B was coarsely pulverized to a size of up to 30 mesh.

[0029]

Subsequently, Alloy A powder and Alloy B powder were weighed in amounts of 90% and 10% by weight, respectively, and mixed together on a V blender for 30 minutes which had been purged with nitrogen. On a jet mill using high-pressure nitrogen gas, the mixed powder was finely pulverized to a mass median particle diameter of 4 μ m. The mixed fine powder was compacted in a nitrogen atmosphere under a pressure of about 1 ton/cm² while being oriented in a magnetic field of 15 kOe. The compact was then placed in a sintering furnace in an argon atmosphere where it was sintered at 1,060°C for 2 hours, obtaining a magnet block dimensioned 10 mm \times 20 mm \times 15 mm (thick). Using a diamond cutter, the magnet block was machined on all the surfaces to 4 mm \times 4 mm \times 0.5 mm (magnetic anisotropy direction).

The machined magnet form was cleaned with an alkaline solution, cleaned with acids and dried. Steps of rinsing with deionized water were included before and after each cleaning step.

[0030]

Subsequently, dysprosium fluoride having an average particle size of 1 μ m was mixed with ethanol at a weight fraction of 50%, in which the magnet form was immersed for one minute with ultrasonic waves being applied. The magnet form was pulled up and immediately dried with hot air. At this point, the filling factor of dysprosium fluoride in the magnet surface-surrounding space was 50%. The magnet form was subjected to absorption treatment in an argon atmosphere at 900°C for one hour, then to aging treatment at 500°C for one hour, and quenched, obtaining a magnet form designated

M1. For comparison purposes, a magnet form designated P1 was prepared by subjecting it to only heat treatment.

[0031]

5 In FIG. 1, demagnetization curves of magnet forms M1 and P1 are depicted as curves H1 and K1, respectively, and their magnetic properties are shown in Table 1. The inventive magnet was found to offer an increase in coercive force of 400 kAm^{-1} relative to the coercive force of the magnet P1 which had not been subjected to dysprosium
10 absorption treatment. A drop of remanence was not found.

[0032]

As Comparative Example 2, a sintered magnet was prepared using an alloy composition in which part of Nd in the above-mentioned Alloy A was substituted with Dy without
15 absorption treatment. This magnet was designed to achieve an increase in coercive force of 400 kAm^{-1} , but its remanence dropped by 40 mT. The magnetic properties of this magnet form P2 are also shown in Table 1.

[0033]

20 [Example 2]

By the same procedure as in Example 1, a magnet form of $4 \text{ mm} \times 4 \text{ mm} \times 0.5 \text{ mm}$ was prepared.

Dysprosium fluoride having an average particle size of $10 \text{ }\mu\text{m}$ was mixed with ethanol at a weight fraction of 50%, in
25 which the magnet form was immersed for one minute with ultrasonic waves being applied. The magnet form was pulled up and immediately dried with hot air. At this point, the filling factor of dysprosium fluoride in the magnet surface-surrounding space was 45% by volume. The magnet
30 form was subjected to absorption treatment in an argon atmosphere at 900°C for one hour, then to aging treatment at 500°C for one hour, and quenched, obtaining a magnet form designated M2.

[0034]

35 The magnetic properties of magnet form M2 are also shown in Table 1. The inventive magnet was found to offer an increase in coercive force of 500 kAm^{-1} relative to the coercive

force of the magnet P1 which had not been subjected to dysprosium absorption treatment. A drop of remanence was 5 mT.

[0035]

Fig. 2 shows the distribution of Dy concentration of the magnet forms M2 and P1 by EPMA (Electron Probe Micro Analysis). In P1, since the magnet alloy as the starting material does not contain Dy, bright contrast which shows the existence of Dy is not found. On the other hand, in the magnet form M2 which is absorption treated by using dysprosium fluoride according to the present invention, Dy is concentrated only in the grain boundary. It was confirmed that the Dy distribution enables the increase of the coercive force while restraining the drop of the remanence to the minimum.

[0036]

15 [Table 1]

		Br (T)	HcJ (kAm ⁻¹)	(BH) _{max} (kJm ⁻³)
Example 1	M1	1.420	1,400	395
Example 2	M2	1.415	1,500	390
Comparative Example 1	P1	1.420	1,000	395
Comparative Example 2	P2	1.380	1,400	375

[BRIEF DESCRIPTION OF THE DRAWINGS]

[0037]

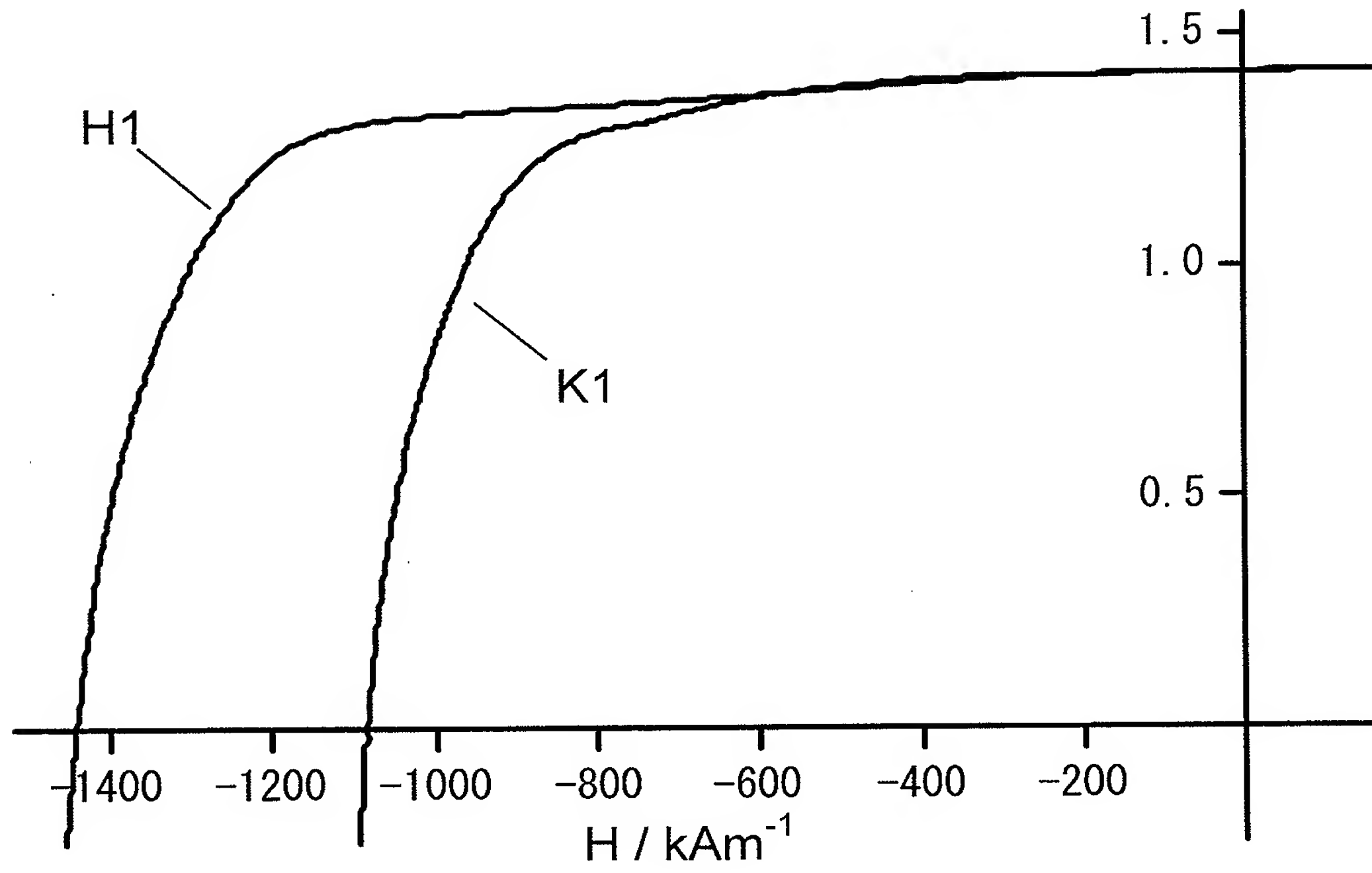
20 [FIG. 1] FIG. 1 is a graph showing a demagnetization curve (curve H1) of a magnet form M1 which is prepared by the invention and a demagnetization curve (curve K1) of a magnet form P1 which is prepared only by machining and heat treatment.

25 [FIG. 2] FIG. 2 is the results of EPMA measurement showing (a) the distribution of Dy concentration of a magnet form M2 which is prepared by the invention and (b) the distribution of Dy concentration of a magnet form P1 which is prepared only by machining and heat treatment.

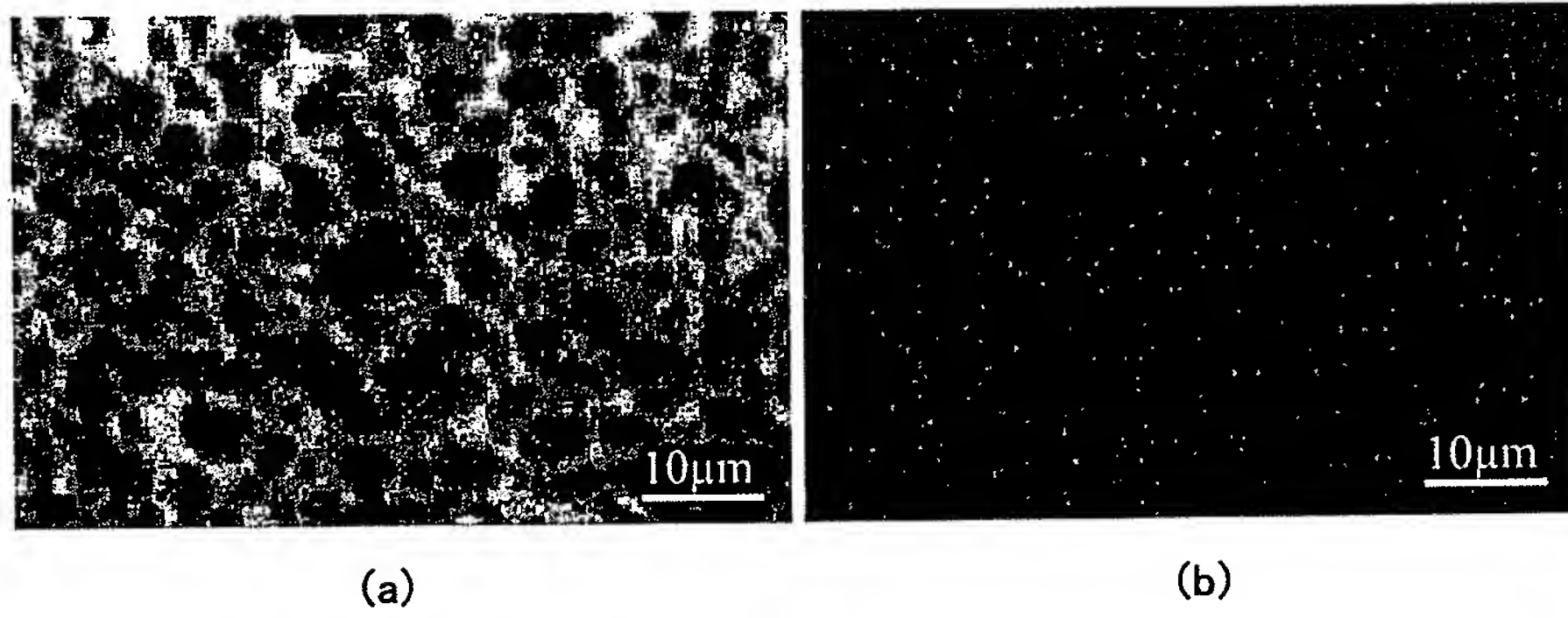
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[DRAWING]

[FIG. 1]



[FIG. 2]



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[ABSTRACT]

[MEANS FOR SOLUTION]

5 A method for preparing a rare earth permanent magnet material comprising the steps of:

forming a sintered magnet form of a R^1 -Fe-B composition wherein R^1 is one or more elements selected from among rare earth elements inclusive of Y and Sc to a shape having a dimension of up to 20 mm along its maximum side and a
10 dimension of up to 2 mm in a magnetic anisotropy direction,

disposing a powder having an average particle size of up to 100 μ m comprising one or more members selected from an oxide of R^2 , a fluoride of R^3 , and an oxyfluoride of R^4 wherein R^2 , R^3 and R^4 each are one or more elements selected
15 from among rare earth elements inclusive of Y and Sc on the sintered magnet form in a magnet-surrounding space within a distance of 1 mm from the surface of the magnet form and at an average filling factor of at least 10%, and

heat treating the magnet form and the powder at a
20 temperature equal to or below the sintering temperature of the magnet in vacuum or in an inert gas for 1 minute to 10 hours, thereby absorbing at least one of R^2 , R^3 and R^4 contained in the powder into the sintered magnet.

[EFFECT]

25 The present invention ensures that a compact or thin permanent magnet having a high remanence and a high coercive force is manufactured at a high productivity.

[SELECTED DRAWING] none